

1 **Lack of correlation between chlorophyll a and cloud droplet effective radius in**
2 **shallow marine clouds**

3

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1 Abstract

2 The hypothesis that areas of high oceanic productivity affect the physical properties of
3 shallow marine clouds via the production of secondary organic aerosols is evaluated
4 using satellite data. The correlation between *chlorophyll a* concentrations, an indication
5 of oceanic productivity, and low cloud droplet liquid phase effective radius (R_e) is
6 examined for several ocean regions and time periods. While a strong correlation between
7 *chlorophyll a* and low R_e can occur for specific periods in some locations, the correlation
8 is not reproducible in other regions and time periods. The intermittent correlation
9 between high concentrations of *chlorophyll a* and low R_e is a coincidence and is not
10 representative of a dominant, monotonic, causative relation between secondary organic
11 aerosols and marine shallow cloud properties.

1 **1 Introduction**

2 Shallow, liquid phase, marine clouds are important to global climate because they
3 reflect a substantial amount of incoming shortwave radiation but have only a small effect
4 on net outgoing longwave radiation compared to the sea surface. The interactions among
5 aerosols, cloud properties, boundary layer dynamics, surface processes, and radiative
6 effects in these shallow marine clouds are complex and can be non-monotonic
7 [Ackerman et al., 2004; Jiang and Feingold, 2006; Xue et al., 2007; Wood, 2007].

8 A 2006 paper by Meskhidze and Nenes (hereafter MN06) explored the effect of
9 oceanic biological productivity on the properties of shallow marine clouds. MN06
10 hypothesized that secondary organic aerosols (SOA), produced as a byproduct of
11 biological production by phytoplankton, can affect the number concentration and size
12 distribution of cloud droplets in marine clouds. They used the concentration of
13 *chlorophyll a*, estimated by the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) on
14 the Seastar satellite, as a proxy for the productivity of phytoplankton. Cloud droplet
15 liquid phase effective radius (R_e), estimated by the Moderate Resolution Imaging
16 Spectroradiometer (MODIS) on the Terra satellite, was monitored to assess any changes
17 in cloud properties. MN06 used the correlation between high concentrations of
18 *chlorophyll a* and low R_e as evidence to support a causative link between oceanic
19 biological productivity and marine cloud characteristics. MN06 argue that increases in
20 *chlorophyll a* concentration lead to increased concentrations of SOA, which in turn cause
21 a decrease in R_e and an increase in cloud droplet concentration. MN06 examined other
22 meteorological controls that could alter R_e and concluded that SOA produced as a

1 byproduct of biological production by phytoplankton blooms is the dominant factor
2 influencing R_e and cloud droplet concentration properties.

3

4 The crux of the MN06 argument relies on the high anticorrelation of *chlorophyll a*
5 to R_e in areas with high *chlorophyll a* concentrations. We tested the reproducibility of this
6 correlation by examining not only MN06's southern Atlantic study region but other
7 oceanic areas that experience phytoplankton blooms. Figure 1 shows the fraction of
8 months from 2001 – 2005 where the concentration of *chlorophyll a* was high enough to
9 indicate strong phytoplankton blooms and the *cloud fraction* was high enough to ensure
10 reliable R_e measurements. *Chlorophyll a* concentrations above 1 mg m^{-3} are considered
11 sufficient to indicate a phytoplankton bloom, and *cloud fraction* values above 0.7 are
12 considered sufficient for reliable R_e measurements. Each of the blue boxes in Fig. 1
13 indicates an area where the spatial correlation between *chlorophyll a* and R_e was
14 evaluated, and the smaller black boxes indicate areas where the time series of *chlorophyll*
15 *a* and R_e were evaluated. The potential effects of *cloud fraction* on the reliability of R_e
16 measurements are discussed in section 5. Table 1 summarizes the spatial correlation
17 statistics for the areas indicated in Fig. 1.

18

19 **2 Spatial Correlation Methodology**

20 To evaluate the spatial correlation between phytoplankton and marine cloud
21 properties, we define the high *chlorophyll a* bloom area as the subset of $1^\circ \times 1^\circ$ grid
22 boxes where the average concentration of *chlorophyll a* was greater than or equal to 1 mg
23 m^{-3} . The remaining grid boxes were considered non-bloom areas. In contrast, MN06

1 defined the latitude band between 48° S and 56° S within the larger geographic area
2 between 55° W to 21° W and 42° S to 60° S as inside the phytoplankton bloom area and
3 the areas to the north and south as outside the bloom. MN06 calculated correlations of
4 oceanic and atmospheric parameters for 2° x 2° grid boxes over their latitude bands as
5 defined above (their Table 1). For the latitude band definition of the bloom area to be
6 applied to other blooms, the size, shape, and geographic surroundings of the bloom would
7 have to be similar to the bloom from the MN06 case.

8

9 The time period 11 December 2001 – 8 January 2002 corresponds to the period
10 studied in MN06 (MN06 Fig. 3a). Using the 1° grid box threshold as opposed to the
11 latitude band definition of bloom and non-bloom areas yields higher correlations between
12 *chlorophyll a* concentration and R_e inside the bloom area (-0.67±0.19-0.13 versus MN06
13 -0.49±0.09) and slightly lower correlations outside the bloom area (-0.02±0.08 versus
14 MN06 0.18±0.14) than were found in MN06. For that event, we agree with MN06 that
15 inside the bloom area there is strong negative correlation between *chlorophyll a*
16 concentrations and R_e (Table 1). The comparable correlations indicate that the 1° grid box
17 threshold methodology used in this study is acceptable for qualitative comparison to the
18 results of the MN06 study.

19

20 **3 Temporal Variation of R_e and Phytoplankton**

21 Figure 2 shows the time series of area-averaged *chlorophyll a* concentration from
22 SeaWIFS monthly products and R_e from MODIS monthly products for two different
23 areas in the southern Atlantic Ocean for the period from March 2000 – December 2005.

1 The area-averaged time series in Fig. 2b is for the same time period and area used in
2 MN06 (their Fig. 2) except that it is compiled from monthly rather than eight-day
3 averaged data. We use monthly data for computational ease since the time series in
4 question are presented for qualitative comparison and analysis.

5

6 For the bloom area in Fig. 2, R_e oscillates on an annual cycle that coincides with
7 the annual phytoplankton blooms. The time series for a nearby area with low *chlorophyll*
8 *a* to the west and upwind of the bloom area is shown in Fig. 2c. Comparison of the time
9 series of R_e for this nearby area with low *chlorophyll a* to the area with high *chlorophyll a*
10 shows that R_e oscillations have similar frequencies and amplitudes regardless of the
11 presence of phytoplankton blooms.

12

13 The Sea of Okhotsk, which lies northeast of Japan, adjacent to Russia's
14 Kamchatka Peninsula (Fig. 3), experiences intense but short-lived phytoplankton blooms
15 during the local spring and summer. During blooms in the Sea of Okhotsk, levels of
16 *chlorophyll a* increase by a factor of ten. As with the area off South Georgia Island [Ward
17 et al., 2005], the phytoplankton blooms in the Sea of Okhotsk are predominantly
18 composed of diatoms [Sorokin and Sorokin, 1999]. This area also has low-cloudiness
19 characteristics similar to the MN06 original study area [Norris, 1998].

20

21 The five-year time series of R_e and *chlorophyll a* concentration for the boxed area
22 in the Sea of Okhotsk shows the annual oscillation in R_e and the local maxima in
23 *chlorophyll a* concentration that denote the annual phytoplankton blooms (Fig. 3). The

1 *chlorophyll a* concentrations and R_e do not have the proper phase relationship to produce
2 a negative correlation that would support the MN06 theory. The minimum in R_e precedes
3 the spike in *chlorophyll a* concentration by ~ 2 months, suggesting that the phenomena
4 which alter the marine cloud properties on an annual basis in the Sea of Okhotsk act
5 before the phytoplankton blooms occur.

6

7 **4 Spatial Correlation of R_e and Phytoplankton**

8 Examination of phytoplankton bloom events across multiple areas facilitates the
9 evaluation of the relationship between phytoplankton and marine cloud properties in a
10 variety of meteorological backgrounds with conditions both similar and different to the
11 southern Atlantic Ocean. Figure 4 shows scatter plots of *chlorophyll a* concentrations and
12 R_e for six bloom events within five geographic areas depicted in the blue boxes in Fig. 1.

13

14 As was stated in section 2, we agree with MN06 that inside the Southern Atlantic
15 bloom area for the time period 11 December 2001 – 8 January 2002, there is strong
16 negative correlation between *chlorophyll a* concentrations and R_e (-0.67+0.19-0.13).
17 However, this relationship is not reproducible in other cases. The correlation between R_e
18 and *chlorophyll a* is weak (-0.1+0.30-0.29) inside the bloom area over the same area that
19 was examined in MN06, but for a bloom from 11 December 2003 – 8 January 2004 of
20 similar size and intensity. The correlation between SeaWiFS estimated *chlorophyll a*
21 concentrations and MODIS estimated R_e for a bloom in the Sea of Okhotsk for 26 June
22 2003 – 27 July 2003 was almost zero (-0.02+0.18-0.17). Furthermore, the spatial
23 correlations from bloom events in the Northwest Atlantic (4 August 2004 – 4 September

1 2004), off the western African Coast (1 January 2002 – 1 February 2002), and in the
2 Bering Sea (7 April 2004 – 8 May 2004) all show a poor relationship between
3 *chlorophyll a* concentrations and R_e (Fig. 4). The bloom and non-bloom area spatial
4 correlations for each case are given in Table 1. With the exception of the bloom event
5 examined in the MN06 study, none of the other events – from different locations and time
6 periods – demonstrate a strong correlation between *chlorophyll a* and R_e in bloom areas.

8 **5 Potential Observational Errors**

9 There are several potential sources of error in satellite estimation of R_e ; chief
10 among them are *cloud fraction* and view angle. The view angle should not represent a
11 source of error that would differentially impact the satellite R_e retrievals since the
12 MODIS satellite's sun-synchronous orbits allows for a constant view angle. Several
13 studies have examined the impacts of *cloud fraction* on the accuracy of MODIS R_e
14 estimations [Coakley et al., 2005; Cornet et al., 2005; Kato et al., 2006]. R_e retrievals are
15 based on 1-D radiative transfer theory that neglects horizontal variations of cloud
16 properties. As a result, R_e is often overestimated. The magnitude of the overestimate is a
17 function of cloud type and decreasing *cloud fraction*. R_e estimates are most reliable where
18 clouds are more homogeneous in the horizontal (e.g. stratus) and less reliable where
19 clouds are less homogeneous in the horizontal (e.g. cumulus).

20
21 Figure 5 shows area-averaged time series of *cloud fraction* and R_e for the three
22 study areas depicted in Fig. 2 and Fig. 3. The area-averaged *cloud fraction* for both the
23 high and low phytoplankton areas in the Southern Atlantic oscillates to a value no lower

1 than 0.84. The *cloud fraction* for the Sea of Okhotsk is more variable with area-averaged
2 *cloud fraction* sometimes dropping just below 0.6 but with a mean above 0.8. If *cloud*
3 *fraction* represented a significant source of error in the MODIS R_e estimations, we would
4 expect to see a local maxima in R_e coinciding with minima in *cloud fraction* representing
5 the bias where low *cloud fraction* leads to overestimation of R_e . Examination of the plots
6 in Fig. 5 illustrates that this is not that case. For the cases examined in this study, *cloud*
7 *fraction* does not appear to represent a significant source of error.

8

9 **6 Conclusions**

10 We have shown that the correlation between low R_e and high *chlorophyll a* shown
11 by MN06 is not reproducible in other regions and time periods (Table 1). MN06's theory
12 that phytoplankton derived SOA is the dominant influence on R_e in marine clouds during
13 plankton blooms requires that high *chlorophyll a* systematically correlate with low R_e .
14 We have shown in Fig. 4 that this is not the case for the southern Atlantic region in
15 December 2003 - January 2004, the Sea of Okhotsk region in June - July 2003, the
16 Northwest Atlantic region in August - September 2004, off the Western African Coast in
17 January 2002, and the Bering Sea region in April - May 2004. The strong correlation
18 between high *chlorophyll a* concentration and low R_e for the area off South Georgia
19 Island for the time period studied by MN06 is a coincidence and not representative of a
20 widespread trend.

21 A recent letter by Wingenter [2007] further questions the conclusions of MN06
22 based on MN06's three-order-of-magnitude overestimation of SOA levels in the
23 phytoplankton bloom area. In their reply, Meskhidze and Nenes [2007] state that this

1 correction does not alter their conclusions because isoprene air-sea fluxes can vary by
2 two orders of magnitude. The isoprene fluxes used in their cloud parcel model
3 simulations represent the high end of measured values within phytoplankton blooms in
4 the Southern Ocean.

5 It is well established that SOA are a source of cloud condensation nuclei (Wallace
6 and Hobbs, 2007). While SOA are likely a contributing factor to shallow marine cloud R_e
7 and number concentration properties, further evidence does not corroborate a direct and
8 strong link between phytoplankton and clouds. The correlation between *chlorophyll a* and
9 R_e is not systematically reproducible and hence is likely a coincidence.

10

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14 406.

1 **Figure Captions**

2 **Fig. 1.** The fraction of months from 2001 – 2005 where *chlorophyll a* concentrations
3 were above 1 mg m^{-3} and *cloud fraction* was above 0.7. The data were averaged to a $1^\circ \times$
4 1° grid. The blue boxes denote the phytoplankton bloom regions examined in this study.
5 The black boxes denote areas where time series were calculated.

6
7 **Fig. 2.** (a) South Atlantic regional map of SeaWiFS-derived *chlorophyll a* concentration
8 derived from monthly data for January 2000 – December 2005. Dashed box indicates
9 area where spatial correlations were calculated. Red box indicates MN06's bloom area,
10 and the white box indicates low-bloom area from which time series in (b) and (c) are
11 respectively derived. White areas denote land or missing data. (b) Time series of monthly
12 *chlorophyll a* concentration and R_e as observed by SeaWiFS and MODIS, respectively,
13 for an area averaged from 49° S to 54° S and 41° W to 35° W (red box). (c) Time series
14 of monthly *chlorophyll a* concentration and R_e as observed by SeaWiFS and MODIS
15 respectively for an area averaged from 52° S to 57° S and 55° W to 49° W (white box).
16

17 **Fig. 3.** (a) Sea of Okhotsk regional map of SeaWiFS-derived *chlorophyll a* concentration
18 derived from 8-day averaged data for a 4-week period for 26 June 2003 – 27 July 2003.
19 The black box indicates the area from which the time series in (b) is derived. White areas
20 denote land or missing data. (b) Time series of monthly *chlorophyll a* concentration and
21 R_e as observed by SeaWiFS and MODIS, respectively, for an area averaged from 57° N
22 to 52° N and 146° E to 152° E .

23

1 **Fig. 4.** Scatter plots of R_e and *chlorophyll a* for bloom (dots) and non-bloom (crosses)
2 areas for the events described in Table 1. The spatial correlation values from Table 1 are
3 shown in upper right of each plot.

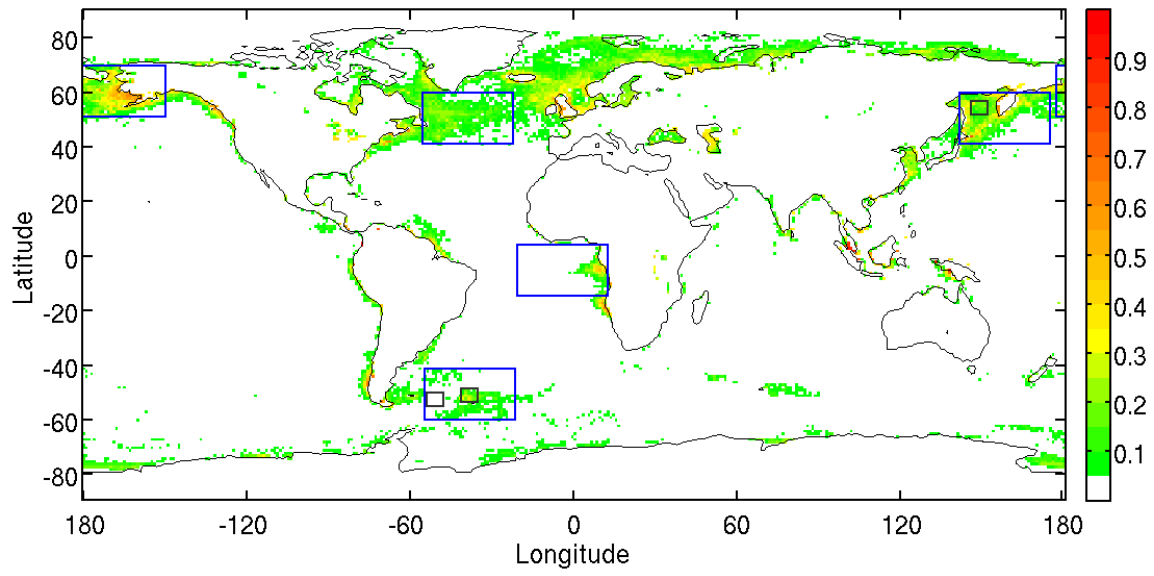
4

5 **Fig. 5.** Area-averaged time series of MODIS *cloud fraction* and R_e from March 2000 –
6 December 2005 for (a) the Southern Atlantic region in the red box in Fig. 2, (b) the
7 Southern Atlantic region in the white box in Fig. 2, and (c) the Sea of Okhotsk region in
8 the black box in Fig. 3.

1 **Table Captions**

2 **Table 1.** The correlations of the concentration of *chlorophyll a* to R_e inside and outside
3 the bloom areas for five different events using the 1° grid box threshold definition of
4 bloom and non-bloom area as outlined in sect. 2. The 95% confidence intervals are given
5 (asymmetry of confidence intervals is a function of correlation and sample size). The
6 correlations for the Northwest Atlantic event are for the area from 42° N to 60° N and 55°
7 W to 21° W. The correlations for the Western African Coast event are for the area from
8 4° N to 14° S and 20° W to 14° E. The correlations for the Bering Sea event are for the
9 area from 52° N to 70° N and 176° E to 150° W.

Fraction of Months From 2001 - 2005 Where Chl. a and CF are Both at High Levels



1

2 **Figure 1**

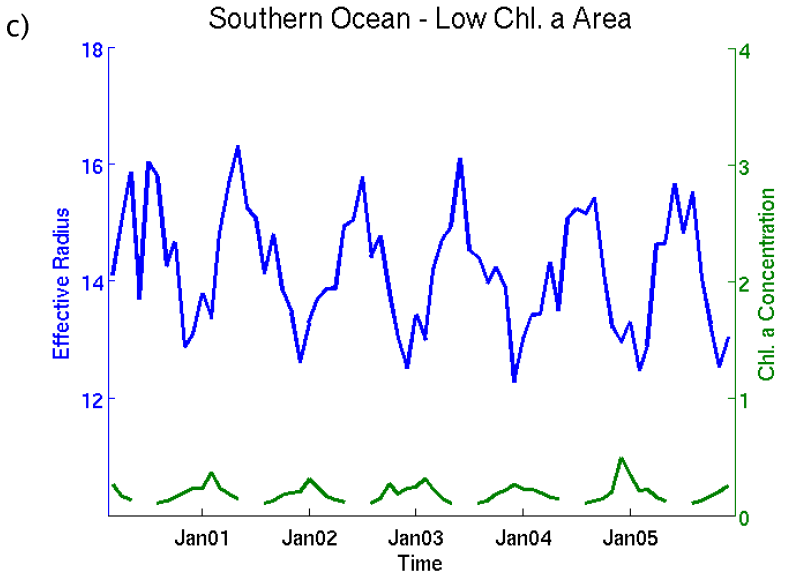
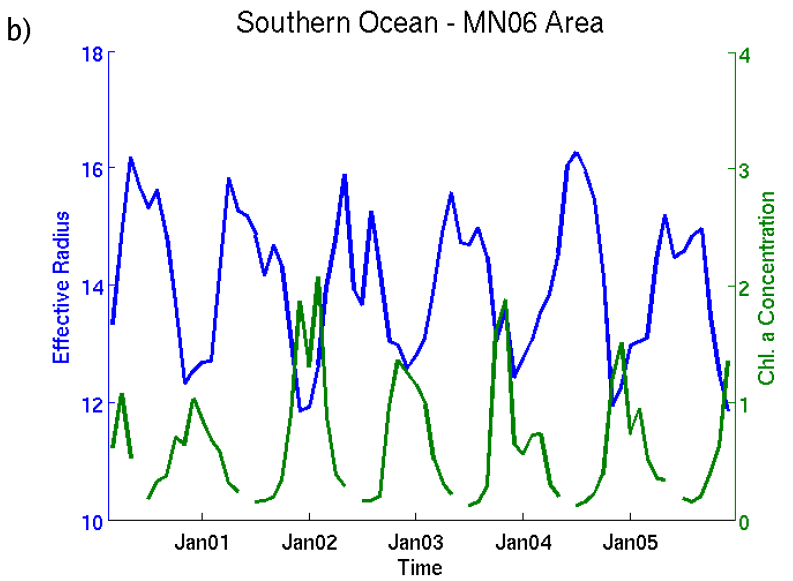
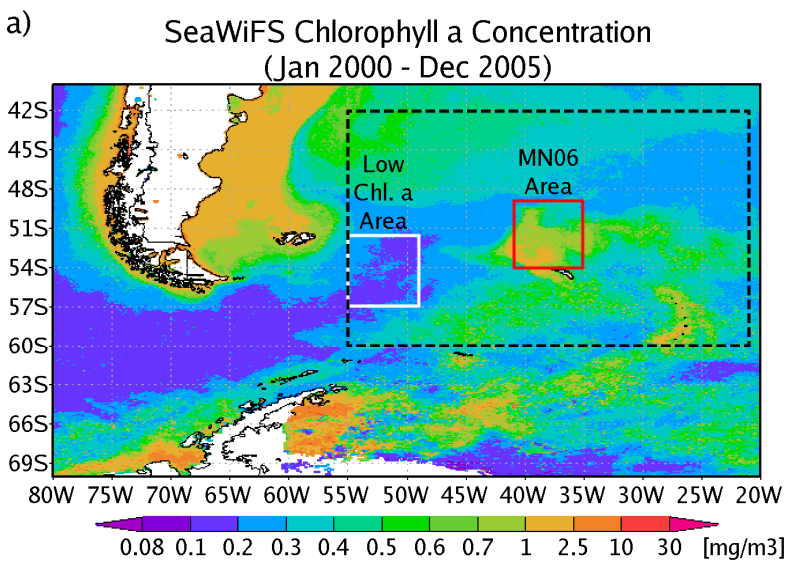
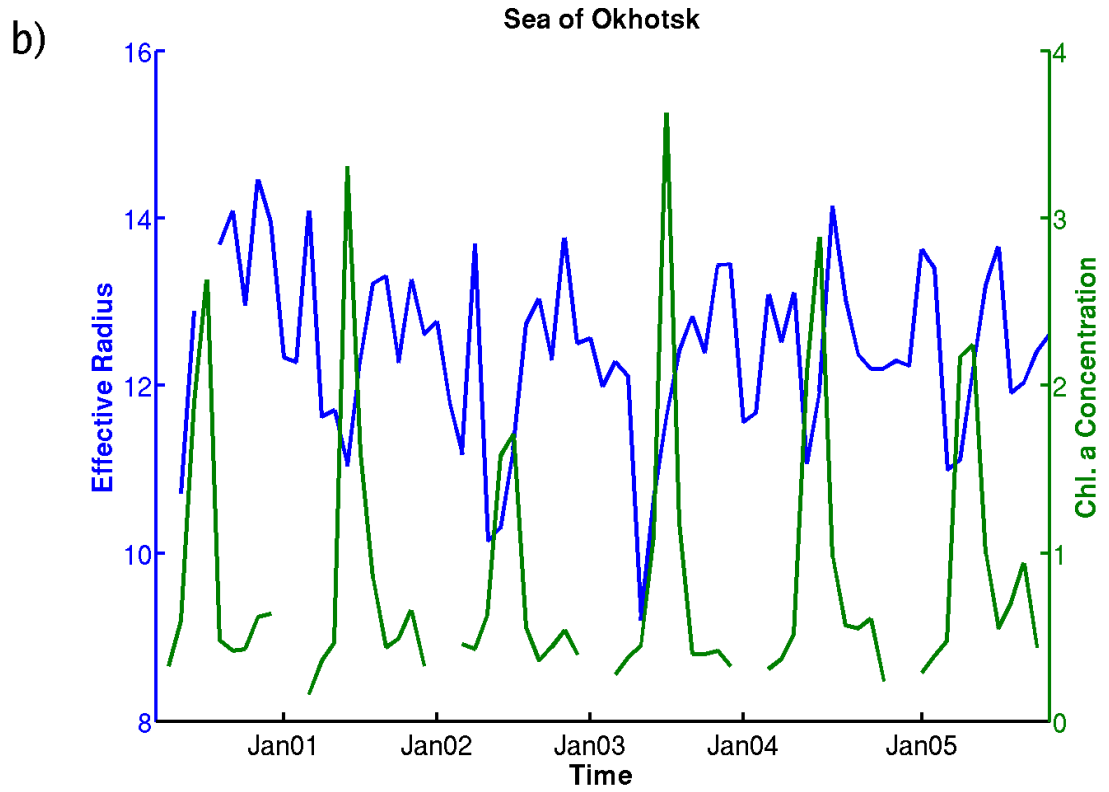
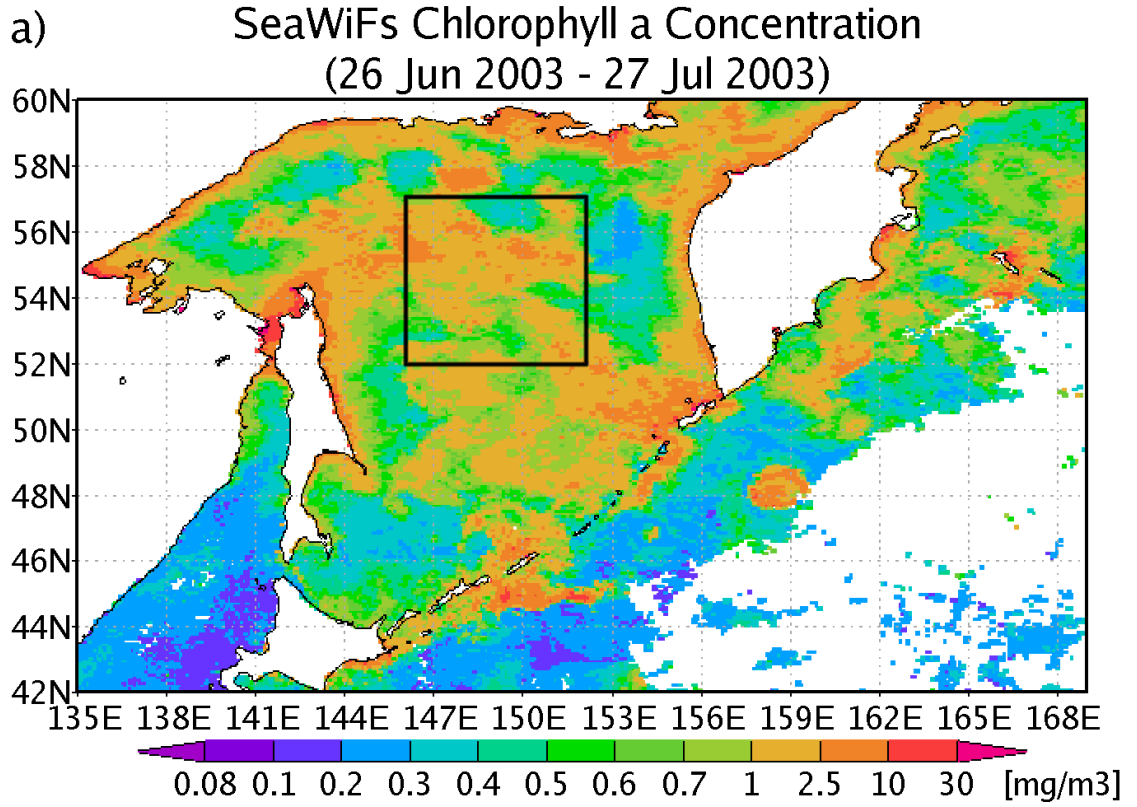
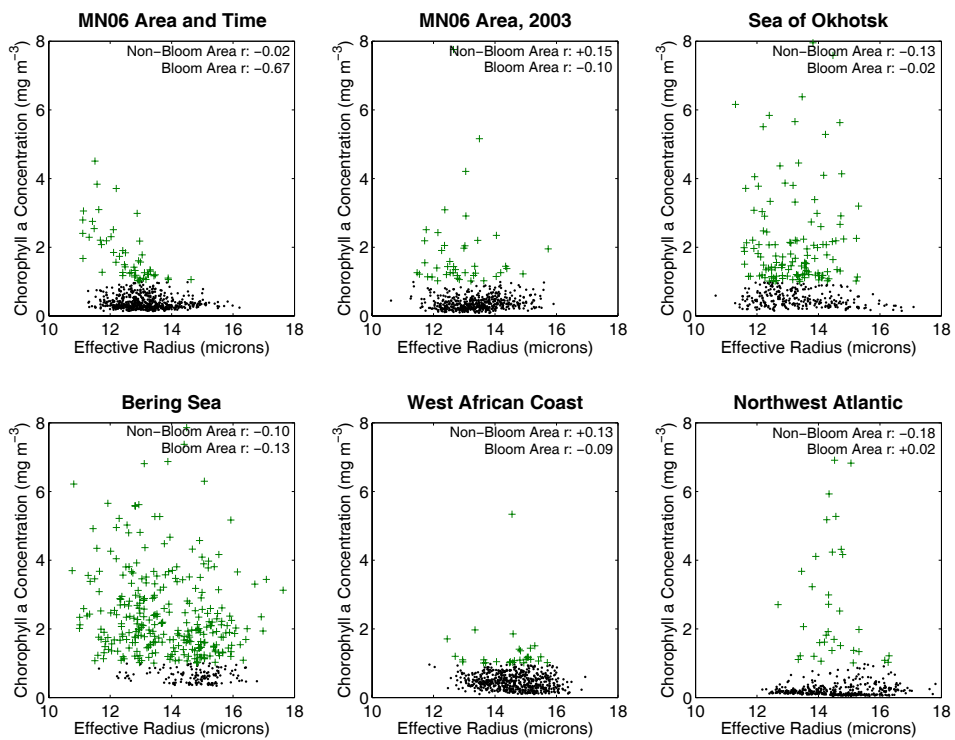


Figure 2



1

2 **Figure 3**



1

2 **Figure 4**

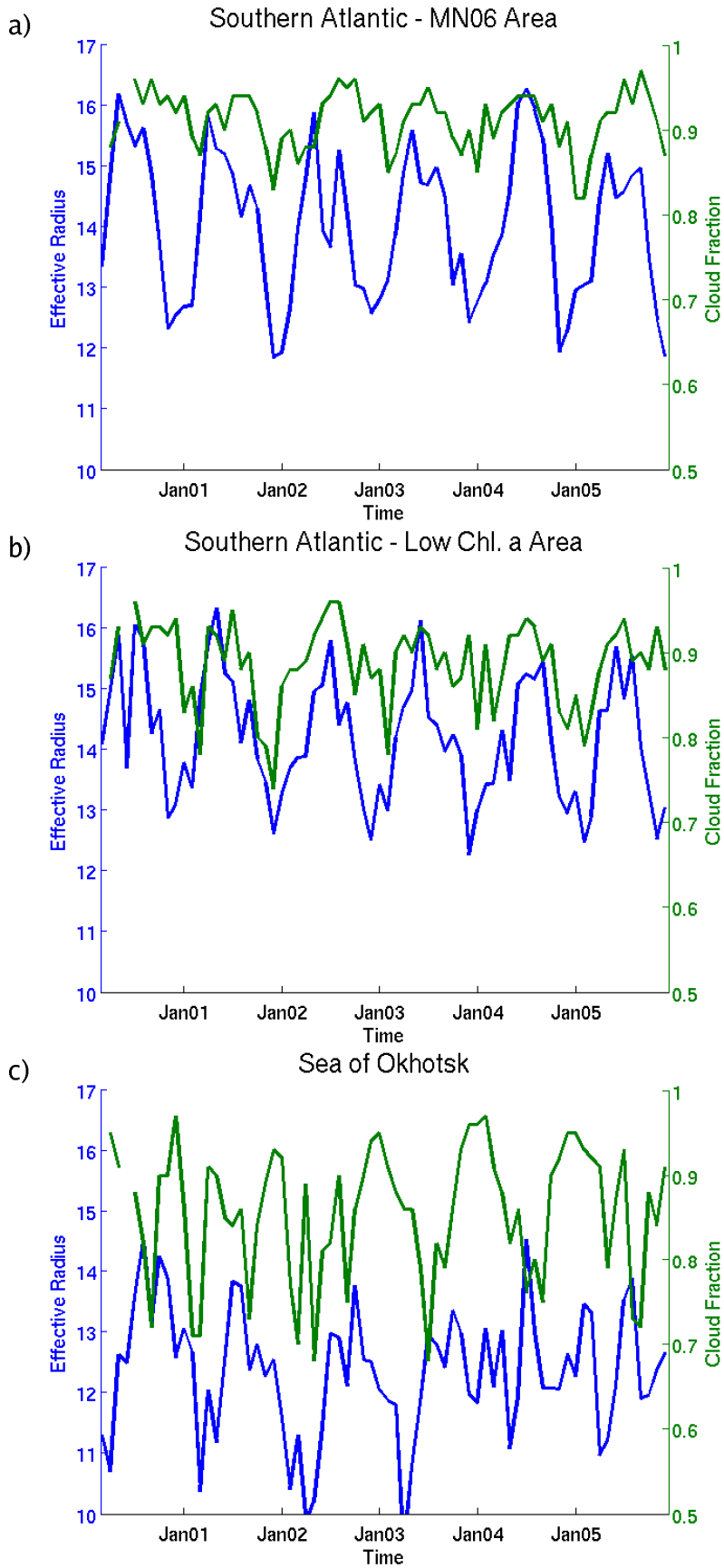


Figure 5

1

Correlation Between *Chlorophyll a* and R_e

Location (Date Range)	Non-Bloom Area <i>Chl. a</i> Concentration (<i>Chl. a</i> < 1 mg m⁻³)	Bloom Area <i>Chl. a</i> Concentration (<i>Chl. a</i> > 1 mg m⁻³)
Southern Atlantic (11 Dec 2001 – 8 Jan 2002)	-0.02±0.08	-0.67 ^{+0.19} _{-0.13}
Southern Atlantic (11 Dec 2003 – 8 Jan 2004)	0.15 ^{+0.08} _{-0.09}	-0.1 ^{+0.30} _{-0.29}
Sea of Okhotsk (26 June 2003 – 27 July 2003)	-0.13±0.12	-0.02 ^{+0.18} _{-0.17}
Northwest Atlantic (4 Aug 2003 – 4 Sep 2004)	-0.18±0.08	-0.09 ^{+0.33} _{-0.31}
West African Coast (1 Jan 2002 – 1 Feb 2002)	0.13 ^{+0.09} _{-0.10}	0.02 ^{+0.36} _{-0.37}
Bering Sea (7 Apr 2004 – 8 May 2004)	-0.1 ^{+0.17} _{-0.16}	-0.13±0.11

2

3 **Table 1**